

Form PTO-1390  
(Rev. 12-29-99)

US DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE

TRANSMITTAL LETTER TO THE UNITED STATES  
DESIGNATED/ELECTED OFFICE (DO/EO/US)  
CONCERNING A FILING UNDER 35 U.S.C. 371ATTORNEY'S DOCKET NO  
**H 4051 PCT/US**U S APPLICATION NO. *If known, see 37 CFR 1.16*  
**10/030851**INTERNATIONAL APPLICATION NO.  
**PCT/EP00/06291**INTERNATIONAL FILING DATE  
**July 5, 2000**PRIORITY DATE CLAIMED  
**July 14, 1999**

## TITLE OF INVENTION

**METHOD FOR TREATING CIRCULATING WATER IN PAINT BOOTHS**

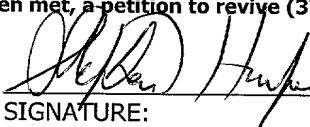
## APPLICANT(S) FOR DO/EO/US

Brigitte Spei, Juergen Geke, Hans-Joergen Rehm and Winfried Wichelhaus

Applicant herewith submits to the United States Designated/Elected Office (EO/DO/US) the following items and other information:

1.  This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2.  This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3.  This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39 (1).
4.  A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.
5.  A copy of the International Application as filed (35 U.S.C. 371(c)(2)).
  - a.  is transmitted herewith (required only if not transmitted by the International Bureau).
  - b.  has been transmitted by the International Bureau.
  - c.  is not required, as the application was filed in the United States Receiving Office (RO/US).
6.  A translation of the International Application into English (35 U.S.C. 371(c)(2)).
7.  Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))
  - a.  are transmitted herewith (required only if not transmitted by the International Bureau).
  - b.  have been transmitted by the International Bureau.
  - c.  have not been made; however, the time limit for making such amendments has NOT expired.
  - d.  have not been made and will not be made.
8.  A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
9.  An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).
10.  A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).
11.  Items 11. to 16. below concern other document(s) or information included:
12.  An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
13.  An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
14.  A FIRST preliminary amendment
15.  A SECOND or SUBSEQUENT preliminary amendment.
16.  A substitute specification
17.  A change of power of attorney and/or address letter.
18.  Other items or information:

International Search Report  
Information Disclosure Citation with References"Express Mail" mailing label number EL615775423US

| U.S. Application No. (If known, see 37 CFR 1.491) <b>10/030851</b>                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                         | INTERNATIONAL APPLICATION NO.<br><b>PCT/EP00/06291</b> | ATTORNEY'S DOCKET NUMBER<br><b>H 4051 PCT/US</b>                                                              |             |        |              |              |      |              |           |   |             |                    |         |   |             |                                              |   |  |            |                                    |  |  |          |
|------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|--------------------------------------------------------|---------------------------------------------------------------------------------------------------------------|-------------|--------|--------------|--------------|------|--------------|-----------|---|-------------|--------------------|---------|---|-------------|----------------------------------------------|---|--|------------|------------------------------------|--|--|----------|
| <p>17. <input checked="" type="checkbox"/> The following fees are submitted:</p> <p><b>BASIC NATIONAL FEE (37 CFR 1.492(a)(1)-(5)):</b></p> <p>Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO..... <b>\$1,000.00</b></p> <p>International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO..... <b>\$890.00</b></p> <p>International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO ..... <b>\$710.00</b></p> <p>International preliminary examination fee paid to USPTO (37 CFR 1.482) but all claims did not satisfy provisions of PCT Article 33(1)-(4) ..... <b>\$690.00</b></p> <p>International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(1)-(4)..... <b>\$100.00</b></p> |                                                        | <b>CALCULATIONS</b> <b>PTO USE ONLY</b>                                                                       |             |        |              |              |      |              |           |   |             |                    |         |   |             |                                              |   |  |            |                                    |  |  |          |
| <b>ENTER APPROPRIATE BASIC FEE AMOUNT</b> =                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                |                                                        | \$ 890                                                                                                        |             |        |              |              |      |              |           |   |             |                    |         |   |             |                                              |   |  |            |                                    |  |  |          |
| Surcharge of <b>\$130.00</b> for furnishing the oath or declaration later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date 37 (CFR 1.492(e)).                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   |                                                        | \$ 0                                                                                                          |             |        |              |              |      |              |           |   |             |                    |         |   |             |                                              |   |  |            |                                    |  |  |          |
| <table border="1" style="width: 100%; border-collapse: collapse;"> <thead> <tr> <th>CLAIMS</th> <th>NUMBER FILED</th> <th>NUMBER EXTRA</th> <th>RATE</th> </tr> </thead> <tbody> <tr> <td>Total Claims</td> <td>28 - 20 =</td> <td>8</td> <td>0 X \$18.00</td> </tr> <tr> <td>Independent Claims</td> <td>4 - 3 =</td> <td>1</td> <td>0 X \$84.00</td> </tr> <tr> <td>Multiple dependent claims (s)(if applicable)</td> <td>0</td> <td></td> <td>+ \$270.00</td> </tr> <tr> <td colspan="3" style="text-align: right;"><b>TOTAL OF ABOVE CALCULATIONS</b></td> <td style="text-align: center;">= \$ 228</td> </tr> </tbody> </table>                                                                                                                                                                                                                                                                                                                                                                                                                       |                                                        |                                                                                                               |             | CLAIMS | NUMBER FILED | NUMBER EXTRA | RATE | Total Claims | 28 - 20 = | 8 | 0 X \$18.00 | Independent Claims | 4 - 3 = | 1 | 0 X \$84.00 | Multiple dependent claims (s)(if applicable) | 0 |  | + \$270.00 | <b>TOTAL OF ABOVE CALCULATIONS</b> |  |  | = \$ 228 |
| CLAIMS                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                     | NUMBER FILED                                           | NUMBER EXTRA                                                                                                  | RATE        |        |              |              |      |              |           |   |             |                    |         |   |             |                                              |   |  |            |                                    |  |  |          |
| Total Claims                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                               | 28 - 20 =                                              | 8                                                                                                             | 0 X \$18.00 |        |              |              |      |              |           |   |             |                    |         |   |             |                                              |   |  |            |                                    |  |  |          |
| Independent Claims                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                         | 4 - 3 =                                                | 1                                                                                                             | 0 X \$84.00 |        |              |              |      |              |           |   |             |                    |         |   |             |                                              |   |  |            |                                    |  |  |          |
| Multiple dependent claims (s)(if applicable)                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                               | 0                                                      |                                                                                                               | + \$270.00  |        |              |              |      |              |           |   |             |                    |         |   |             |                                              |   |  |            |                                    |  |  |          |
| <b>TOTAL OF ABOVE CALCULATIONS</b>                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                         |                                                        |                                                                                                               | = \$ 228    |        |              |              |      |              |           |   |             |                    |         |   |             |                                              |   |  |            |                                    |  |  |          |
| Reduction of $\frac{1}{2}$ for filing by small entity, if applicable. A Small Entity Statement must also be filed. (Note 37 CFR 1.9, 1.27, 1.28).                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                          |                                                        | \$ 0                                                                                                          |             |        |              |              |      |              |           |   |             |                    |         |   |             |                                              |   |  |            |                                    |  |  |          |
| <b>SUBTOTAL</b> =                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                          |                                                        | \$ 1118                                                                                                       |             |        |              |              |      |              |           |   |             |                    |         |   |             |                                              |   |  |            |                                    |  |  |          |
| Processing fee of <b>\$130.00</b> for furnishing the English translation later the <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(f)).                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                               |                                                        | \$ 0                                                                                                          |             |        |              |              |      |              |           |   |             |                    |         |   |             |                                              |   |  |            |                                    |  |  |          |
| <b>TOTAL NATIONAL FEE</b> =                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                |                                                        | \$ 1118                                                                                                       |             |        |              |              |      |              |           |   |             |                    |         |   |             |                                              |   |  |            |                                    |  |  |          |
| Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                     |                                                        | \$ 0                                                                                                          |             |        |              |              |      |              |           |   |             |                    |         |   |             |                                              |   |  |            |                                    |  |  |          |
| <b>TOTAL FEES ENCLOSED</b> =                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                               |                                                        | \$ 1118                                                                                                       |             |        |              |              |      |              |           |   |             |                    |         |   |             |                                              |   |  |            |                                    |  |  |          |
| Amount to be:<br>refunded:                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                 |                                                        | \$ -----                                                                                                      |             |        |              |              |      |              |           |   |             |                    |         |   |             |                                              |   |  |            |                                    |  |  |          |
| charged:                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   |                                                        | \$ 1118.00                                                                                                    |             |        |              |              |      |              |           |   |             |                    |         |   |             |                                              |   |  |            |                                    |  |  |          |
| <p>a. <input type="checkbox"/> A check in the amount of \$_____ to cover the above fees is enclosed.</p> <p>b. <input checked="" type="checkbox"/> Please charge my Deposit Account No. <u>01-1250</u> in the amount of <u>\$1118.00</u> to cover the above fees. A triplicate copy of this sheet is enclosed. Order No. <u>02-0018</u>.</p> <p>c. <input checked="" type="checkbox"/> The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. <u>01-1250</u>. A duplicate copy of this sheet is enclosed.</p> <p><b>NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137 (a) or (b)) must be filed and granted to restore the application to pending status.</b></p>                                                                                                                                                                                                                                |                                                        |                                                                                                               |             |        |              |              |      |              |           |   |             |                    |         |   |             |                                              |   |  |            |                                    |  |  |          |
| SEND ALL CORRESPONDENCE TO:                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                |                                                        | Henkel Corporation, Law Dept.<br>2500 Renaissance Blvd., Suite 200<br>Gulph Mills, PA 19406                   |             |        |              |              |      |              |           |   |             |                    |         |   |             |                                              |   |  |            |                                    |  |  |          |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                            |                                                        | <br><b>SIGNATURE:</b>     |             |        |              |              |      |              |           |   |             |                    |         |   |             |                                              |   |  |            |                                    |  |  |          |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                            |                                                        | <b>Stephen D. Harper</b><br><b>NAME ATTORNEY FOR APPLICANT</b><br><b>33,243</b><br><b>REGISTRATION NUMBER</b> |             |        |              |              |      |              |           |   |             |                    |         |   |             |                                              |   |  |            |                                    |  |  |          |

Express Mail No. EL 615775423US

PATENT  
Docket No. H 4051 PCT/US

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

RE: PCT/EP00/06291  
International Filing Date: July 5, 2000  
Priority Date Claimed: July 14, 1999  
Applicant: Spei et al.  
Title: METHOD FOR TREATING CIRCULATING WATER IN PAINT  
BOOTHS

**PRELIMINARY AMENDMENT**

Assistant Commissioner for Patents  
Box PCT  
Washington, DC 20231

ATTN: DO/EO/US

Sir:

Prior to substantive examination of the above-referenced application, please enter the following Preliminary Amendment.

**IN THE SPECIFICATION:**

Page 1, after the title, insert: --This application claims priority from German Application 19932766.1, filed July 14, 1999, and International Application PCT/EP00/06291 (published in a language other than English), filed July 5, 2000.--

Enter page 14 of the specification enclosed herewith as the Abstract of the Disclosure.

**IN THE CLAIMS:**

Cancel claims 1-14 without prejudice.

Enter the following new claims 15-42:

15. (New) A process for treating circulating water in a painting booth wherein the circulating water is comprised of paint particles, said process comprising adding to the circulating water at least one dispersant selected from the group consisting of:

- a) nonionic surfactants which are alkoxylates of fatty acids and fatty amines;  
and
- b) anionic surfactants;

in a total dispersant concentration of 0.01 to 2.0% by weight, based on circulating water, subject to the proviso that when an anionic surfactant is present no polyaspartic acid

is added to the circulating water.

16. (New) The process of claim 15 wherein at least one dispersant selected from the group consisting of nonionic surfactants which are alkoxylates of fatty acids and fatty amines, said fatty acids and fatty amines containing fatty alkyl groups of 7 to 36 carbon atoms, is added to the circulating water.

17. (New) The process of claim 15 wherein at least one dispersant selected from the group consisting of nonionic surfactants which are alkoxylates of fatty acids and fatty amines, said alkoxylates containing 5 to 100 alkylene oxide units, is added to the circulating water.

18. (New) The process of claim 15 wherein at least one dispersant which is an anionic surfactant selected from the group consisting of soaps, alkyl sulfates, alkyl sulfonates, alkyl benzene sulfonates, alkyl ether sulfates containing alkyl groups having 7 to 44 carbon atoms in said alkyl groups, and sulfonated maleic acid esters is added to the circulating water.

19. (New) The process of claim 15 comprising an additional step of removing the paint particles from the circulating water by membrane filtration.

20. (New) A process for treating circulating water in a painting booth wherein the circulating water is comprised of paint particles, said process comprising adding to the circulating water i) at least one dispersant selected from the group consisting of:

- a) homopolymers and copolymers prepared by polymerization of at least one polymerizable monomer selected from acrylic acid or methacrylic acid, said homopolymers and copolymers having molecular weights of from 2,500 to 500,000;
- b) nonionic surfactants;
- c) inorganic complexing agents; and
- d) nonpolymeric organic complexing agents;

and ii) 0.2 to 2% by weight, based on circulating water, of polyaspartic acid.

21. (New) The process of claim 20 comprising an additional step of removing the paint particles from the circulating water by membrane filtration.

22. (New) The process of claim 20 wherein at least one dispersant selected from the group consisting of homopolymers and copolymers prepared by polymerization of at

Preliminary Amendment of U.S. National Stage for International Application  
PCT/EP00/06291 filed July 5, 2000

least one polymerizable monomer selected from acrylic acid or methacrylic acid, said homopolymers and copolymers having molecular weights of from 15,000 to 250,000, is added to the circulating water.

23. (New) A process for treating circulating water in a painting booth wherein the circulating water is comprised of paint particles and has a hardness of at least 2° German hardness, said process comprising adding to the circulating water at least one dispersant in a total dispersant concentration of 0.01 to 2.0% by weight, based on circulating water, so as to achieve a mean particle size for the paint particles of less than 20 µm, as determined by laser diffraction.

24. (New) The process of claim 23 wherein at least one dispersant is selected from the group consisting of:

- a) homopolymers and copolymers prepared by polymerization of at least one polymerizable monomer selected from acrylic acid or methacrylic acid, said homopolymers and copolymers having molecular weights of from 2,500 to 500,000;
- b) nonionic surfactants;
- c) inorganic complexing agents; and
- d) nonpolymeric organic complexing agents.

25. (New) The process of claim 23 wherein at least one dispersant is a nonionic surfactant selected from the group consisting of alkoxylates of fatty acids, alkoxylates of fatty alcohols and alkoxylates of fatty amines, wherein the nonionic surfactant contains a fatty alkyl group having 7 to 36 carbon atoms and 5 to 100 alkylene oxide units.

26. (New) The process of claim 23 wherein at least one dispersant is an inorganic complexing agent selected from the group consisting of oligomeric and polymeric inorganic phosphates.

27. (New) The process of claim 23 wherein at least one dispersant is a nonpolymeric organic complexing agent selected from the group consisting of organic carboxylic acids containing two to ten hetero atoms capable of co-ordination to metal ions and organic phosphonic acids.

28. (New) The process of claim 23 wherein at least one dispersant is selected from

the group consisting of citric acid, tartaric acid, malic acid, gluconic acid, nitriloacetic acid, ethylenediamine tetraacetic acid, methylglycine diacetic acid, 1-hydroxyethane-1, 1-diphosphonic acid, aminotrimethylene phosphonic acid, phosphonobutane tricarboxylic acid, Na triphosphate, Na pyrophosphate and Na hexametaphosphate.

29. (New) The process of claim 23 wherein 0.2 to 2% by weight, based on the circulating water, of polyaspartic acid is additionally added to the circulating water.

30. (New) The process of claim 23 wherein at least one dispersant is an anionic surfactant and no polyaspartic acid is added to the circulating water.

31. (New) The process of claim 30 wherein the anionic surfactant is selected from the group consisting of soaps, alkyl sulfates, alkyl sulfonates, alkyl benzene sulfonates, alkyl ether sulfates containing alkyl groups having 7 to 44 carbon atoms, and sulfonated maleic acid esters.

32. (New) The process of claim 23 comprising an additional step of removing the paint particles from the circulating water by membrane filtration.

33. (New) A composition comprised of

- a) water;
- b) dispersed paint particles;
- c) polyaspartic acid in acid form, in salt form or in both acid and salt form;
- d) an alkoxylate of a fatty amine; and
- e) an organic phosphonic acid in acid form, in salt form or in both acid and salt form.

34. (New) The composition of claim 33 additionally comprising an alkali metal hydroxide.

35. (New) The composition of claim 33 wherein the organic phosphonic acid is selected from the group consisting of 1-hydroxyethane-1, 1-diphosphonic acid, aminotrimethylene phosphonic acid, and phosphonobutane tricarboxylic acid.

36. (New) The composition of claim 33 wherein said fatty amine is a coconut amine.

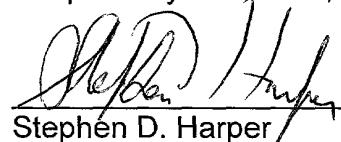
37. (New) The composition of claim 33 wherein said alkoxylate of said fatty amine contains ethylene oxide units.

38. (New) The composition of claim 33 wherein d) and e) are present in a concentration totaling from 0.01 to 2.0% by weight.

Preliminary Amendment of U.S. National Stage for International Application  
PCT/EP00/06291 filed July 5, 2000

39. (New) The composition of claim 33, wherein the polyaspartic acid is present at a concentration of 0.2 to 2% by weight.
40. (New) The composition of claim 33 wherein said dispersed paint particles have an average particle size of less than 20  $\mu\text{m}$ .
41. (New) The composition of claim 33 wherein said composition has a weakly alkaline pH.
42. (New) The composition of claim 33 wherein said composition has a pH of from about 8 to about 10.

Respectfully submitted,

  
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Henkel KGaA - H 4051  
Dr. Endres / KK  
13.07.1999

- 1 -

"Process for the treatment of the circulating water in painting booths"

This invention relates to a process for the treatment of the circulating water in painting booths, particularly for vehicle painting. The paint particles washed out of the waste air and introduced into the circulating water are kept in suspension in finely dispersed form and removed from the circulating water continuously or discontinuously. This is achieved by addition of suitable additives to the circulating water, the concentration of which in the circulating water is set to the relatively low values of between 0.01 and 0.5 wt. % based on the circulating water.

In painting booths in which water-based or solvent-based paints are applied to the surfaces to be painted, the entire quantity of paint used is not applied to the intended surface. Rather, a considerable proportion of the paint particles becomes mixed with the air flowing through the painting booth or settles on the walls of the painting booth. The paint particles, the so-called paint overspray, are washed out of the air stream with water by means of suitable washing devices, such as Venturi scrubbers.

The water mixed with paint is collected in circulating water tanks and circulated. As the paint particles in the circulating water stick together and to equipment parts, however, they are released and coagulated by traditional methods by the addition of release and coagulation agents. The coagulated paint particles settle or float on the circulating water according to the method employed. The thus-produced paint sludge may be removed from the bottom or the surface of the circulating water tank by means of suitable removal devices. This traditional method requires the use of inorganic or organic release and coagulation agents which may be selected, for example, from sheet silicates, waxes and organic polymers. The quantity used of these products is generally from 10 to 50 wt. % based on the paint overspray ready for spraying. The release of the paint particles and removal of the paint sludge is not, however, usually complete so that paint sludge is deposited and has to be removed by hand from time to time, involving a great deal of work. In addition, an additional quantity of sludge is produced where inorganic coagulants are used, and this leads to additional disposal costs.

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There is therefore a need for improved processes for the treatment of the circulating water in painting booths, which may involve a reduced use of chemicals, with which fewer problems caused by sticky paint particles arise and/or in which there is less manual outlay for cleaning and maintaining the painting booths and the disposal costs are distinctly minimized.

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In a first embodiment, the present invention provides a process for the treatment of the circulating water in painting booths, wherein the paint particles are dispersed by addition of dispersants which are selected from:

- (a) homo- and co-polymers based on maleic acid, acrylic acid and/or methacrylic acid having molecular weights of between 2,500 and 500,000;
- 10 (b) non-ionic surfactants;
- (c) anionic surfactants, no polyaspartic acid being additionally added to the circulating water in this case;
- (d) inorganic or non-polymeric organic complexing agents and mixtures thereof;
- 15 the total concentration thereof being between 0.01 and 2.0 wt. %, preferably between 0.02 and 0.5 wt. %, based on the circulating water.

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The success of the process according to the present invention is based on the fact that the paint particles remain dispersed in the circulating water and are not deposited as paint sludge which is difficult to remove. The circulating water may therefore be circulated together with the paint particles dispersed therein without equipment parts becoming sticky.

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Homo- and co-polymers based on maleic acid, acrylic acid and/or methacrylic acid are intended to mean those polymers wherein at least part of the monomers consists of the above-mentioned polymerizable carboxylic acids. Other polymerizable monomers may be co-incorporated into the polymers. Polymers which exclusively contain acrylic acid and/or methacrylic acid are used, for example. The molecular weight of the polymers is preferably from 15,000 to 250,000, particularly up to 50,000. The polymers may be added to the circulating water as they are or in salt

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form. Depending on the pH of the circulating water, the acid/base equilibrium between acid and salt form is automatically adjusted.

In the process according to the present invention, non-ionic surfactants, which are preferably selected from alkoxylates of fatty acids, fatty alcohols or fatty amines having 7 to 36, preferably 10 to 22, carbon atoms in the fatty alkyl group and having 5 to 100, preferably 10 to 80, alkylene oxide units, may be used as dispersants. These alkoxylates may have the terminal groups thereof closed, i.e. represent so-called mixed ethers.

A butyl group may be present, for example, as a terminal group. Ethylene oxide groups or combinations of ethylene oxide groups and propylene oxide groups are considered in particular as alkylene oxide groups. An alkoxyate of a C<sub>12/14</sub>-fatty alcohol mixture with an average of five ethylene oxide and four propylene oxide units in the molecule may be used for example.

Inorganic or non-polymeric organic complexing agents may further be used as dispersants. These are preferably selected from:

- (i) organic carboxylic acids having two to ten heteroatoms, which may coordinate on metal ions, particularly from citric acid, tartaric acid, malic acid, gluconic acid, nitrilotriacetic acid, ethylenediamine tetraacetic acid, methylglycine diacetic acid;
- (ii) organic phosphonic acids, particularly from 1-hydroxyethane-1,1-diphosphonic acid, aminotrimethylene phosphonic acid and phosphonobutane tricarboxylic acid;
- (iii) oligomeric or polymeric inorganic phosphates, particularly Na triphosphate, Na pyrophosphate and Na hexametaphosphate.

Oxygen atoms (alcohols, carboxylates) or nitrogen atoms are considered in particular as heteroatoms of the organic complexing agents which may coordinate on metal ions.

It is also applicable in each case that the complexing agents may be used in the form of the acids or in salt form. Depending on the pH of the circulating water, a mixture comprising acid and salt form will be reached according to the acid/base equilibrium.

The organic complexing agents should not be polymers and should differ in this way from the polymeric carboxylic acids of (a), which also have complexing properties. The inorganic complexing agents may, however, be entirely polymeric, such as Na pyrophosphate.

5 Mixtures of representatives of the individual groups of dispersants listed may also be used. Mixtures comprising polyacrylates and phosphonic acids and mixtures comprising niosurfactants and phosphonic acids are particularly suitable.

Where dispersants selected from (a), (b) and (d) are used, 0.2 to 2 wt. %, based on the circulating water, of polyaspartic acid may be additionally added to the circulating water. The use of polyaspartic acid as such is known from patent application PCT/EP99/00350, which is not a prior publication. According to this document, the polyaspartic acid may be used together with anionic surfactants. In the process according to the present invention, however, anionic surfactants may also be used without the co-use of polyaspartic acid. The anionic surfactants to be used as dispersants are preferably selected from soaps, alkyl sulfates, alkyl sulfonates, alkylbenzene sulfonates, alkylether sulfates each having 7 to 44, preferably 8 to 22, carbon atoms in the alkyl group, and from sulfonated maleic acid esters.

20 In the process according to the present invention, the pH of the circulating water is preferably adjusted to a range between about 5 and about 10.5. A weakly alkaline pH may be preferred. Particularly where phosphonic acids are used as dispersants, particularly advantageous results are obtained when the pH in the circulating water is between about 8 and about 10. At such a pH, dispersants capable of protolysis will be mainly present in salt form, irrespective of the form in which they are introduced into the circulating water. Depending on the form in which the dispersants are used, it may be necessary additionally to use pH adjusters, such as alkali metal hydroxides, to give the preferred pH range.

25 Depending on the type of paint used, it may further be advisable additionally to add defoaming agents to the circulating water. The use of defoaming agents is generally

known in the technical field in question as foam problems may arise in the circulating water particularly where water-based paints are used.

5 The process according to the present invention is particularly suitable for painting equipment in which the circulating water exhibits high turbulence. The dispersing

effect of the dispersants to be used according to the present invention is increased by

the flowing of the circulating water.

When the dispersants according to the present invention are used, the circulating water may be circulated for a certain period of time without the need to remove the entrained paint particles. If the equipment is operated for a fairly long time,

however, as time progresses a concentration of paint particles in the circulating water is reached which should not be further exceeded in order to prevent breakdowns. The range in which this concentration limit lies depends on the actual design of the equipment in question. For this reason, the process according to the present invention preferably provides for the removal of a proportion of the paint particles

10 from the circulating water continuously or discontinuously. The preferred procedure in this case is to separate some of the paint particles from the circulating water by means of a membrane filtration, preferably an ultra filtration. The permeate of the membrane filtration is returned to the circulating water and the retentate disposed of.

15 For this purpose, a portion of the circulating water may be passed over the membrane via a by-pass discontinuously or continuously.

20

The process according to the present invention functions particularly reliably when it may be ensured, by addition of the dispersants to the circulating water in a concentration between 0.01 and 2.0 wt. % based on the circulating water, that the paint particles in the circulating water have an average particle size, which may be

25 determined by means of laser diffraction, of below about 20  $\mu\text{m}$ . To achieve this, an addition of suitable dispersants is required for circulating water of a hardness of at least 2 °dH (German hardness) (corresponds to 0.714 mval/l of alkaline earth metal ions).

Accordingly, in a generalized aspect, the present invention relates to a process for the treatment of the circulating water in painting booths wherein the circulating water has a hardness of at least 2° German hardness and wherein the paint particles are dispersed by addition of dispersants in a concentration between 0.01 and 2.0 wt. % based on the circulating water in such a way that they exhibit an average particle size, which may be determined by means of laser diffraction, of below 20  $\mu\text{m}$ .

Laser diffraction is an example of a method of measurement of particle sizes in the micrometer range, in which the light scatter brought about by the particles is measured. A particle size measuring instrument made by Sympatec may be used for this purpose, for example. The average particle size is defined as that particle size which establishes when the particles are kept in suspension by a slight movement of the suspension, for example, by stirring. Ultrasound, which mechanically destroys the particles, is not used in this case.

The above statements apply to the dispersants which may be used under this generalized aspect of the present invention. This also applies to the preferred procedure described above.

In the two embodiments expressed by the independent claims, the present invention is suitable for dispersing water-based and/or solvent-based paints.

**Examples**

Herberts' water-based metallic paint Aqua Pearl Base, Siena red II was used for the trials. In each case, a quantity of test paint was introduced into water so that the paint solids content was 0.5 wt. % based on the amount of water. Düsseldorf/Benrath 5 tap water, which has a hardness of 18 °dH (corresponding to 6.426 mval/l of alkaline earth metal ions) was used as the water.

In a first set of trials, Sympatec's particle size measuring instrument based on laser 10 diffraction was used to measure the average particle size (defined as 50% value) which was established under gentle stirring using the various dispersants. The results are contained in Table 1.

**Table 1: Average particle size (50% value)**

| Example   | Dispersant/quantity with respect to water                                    | 50% value (μm) |
|-----------|------------------------------------------------------------------------------|----------------|
| Ref. 1    | none                                                                         | 255            |
| Example 1 | Coconut amine x 12 EO <sup>(1)</sup> , 0.25 %                                | 11.8           |
| Example 2 | Coconut amine x 12 EO <sup>(1)</sup> , 0.25 %<br>+ polyaspartic acid, 0.76 % | 11.3           |

<sup>(1)</sup> EO = ethylene oxide

In the above set of trials, it was shown that the particle size correlates to the stability 20 of the paint suspension: at a particle size below about 20 μm, no sedimentation or flotation may be observed, even 30 minutes after the dispersion was prepared. Paint does not settle on the glass wall of the test vessel. Further dispersants were therefore only then tested to see whether they exhibit this behavior. Table 2 contains examples.

Table 2: Dispersibility: dispersion stability after 30 minutes

| Ex. No. | Dispersant quantity (wt. % based on water) | Dispersion stable?                                        | Paint on vessel wall? |
|---------|--------------------------------------------|-----------------------------------------------------------|-----------------------|
| 5       | Ref. 2                                     | none                                                      | no (flootation)       |
|         | Ref. 3                                     | polyaspartic acid, 0.76%                                  | no (flootation)       |
|         | Ex. 3                                      | modified polyacrylic acid<br>molecular weight 4000, 0.23% | yes                   |
|         | Ex. 4                                      | coconut amine x 12 EO                                     | yes                   |
|         | Ex. 5                                      | sulfosuccinic acid-bis-2-ethylhexylester, Na salt, 0.38%  | yes                   |
|         | Ex. 6                                      | Na oleate, 0.5%                                           | yes                   |

10 The sediment volume was quantitatively determined in a further set of trials. For this purpose, the dispersant was placed in a liter of water and stirred for 10 minutes at 300 rpm. The test paint described above was then added in a quantity such that the paint solids content was 0.5 wt. % based on the water. Stirring was repeated for 10 minutes at 300 rpm. The dispersion was then transferred into a measuring funnel (Imhoff cone) and the sediment volume, in ml, measured after a residence time of 10 minutes. Good dispersion is evident from the fact that no sediment appears. Table 15 3 contains the results.

Table 3: Sediment volume

| Ex. No.                                                                   | Dispersant quantity (wt. % based on water)          | Sediment volume (ml) |
|---------------------------------------------------------------------------|-----------------------------------------------------|----------------------|
| Ref. 4                                                                    | none                                                | 100                  |
| Ref. 5                                                                    | polyaspartic acid, 0.2%                             | 2.5                  |
| 5                                                                         | Ex. 7 polyacrylate (molecular weight 10,000), 0.18% | 0                    |
| Ex. 8 maleic acid/olefin copolymer (molecular weight 12,000), 0.13%       | 0                                                   |                      |
| Ex. 9 polyacrylate (molecular weight 30,000), 0.2%                        | 0                                                   |                      |
| Ex. 10 maleic acid/acrylic acid copolymer (molecular weight 70,000), 0.2% | 0                                                   |                      |
| Ex. 11 coconut amine x 12 EO, 0.1%                                        | 0                                                   |                      |
| 10                                                                        | Ex. 12 Na tripolyphosphate, 0.5%                    | 0                    |
| Ex. 13 Na pyrophosphate, 0.5%                                             | 0                                                   |                      |
| Ex. 14 Na hexametaphosphate, 0.5%                                         | 0                                                   |                      |
| Ex. 15 citric acid, 0.5%                                                  | 0                                                   |                      |
| Ex. 16 1-hydroxyethane-1,1-diphosphonic acid, 0.3%                        | 0                                                   |                      |
| 15                                                                        | Ex. 17 aminotrimethylene phosphonic acid, 0.25%     | 0                    |
| Ex. 18 phosphonobutane tricarboxylic acid, 0.25%                          | 0                                                   |                      |
| Ex. 19 nitrilotriacetic acid, 0.2%                                        | 0                                                   |                      |
| Ex. 20 C <sub>16/18</sub> -fatty alcohol x 25 EO, 0.5%                    | 0                                                   |                      |
| Ex. 21 C <sub>16/18</sub> -fatty alcohol x 80 EO, 0.5%                    | 0                                                   |                      |
| 20                                                                        | Ex. 22 coconut amine x 15 EO, 0.5%                  | 0                    |
| Ex. 23 C <sub>12/14</sub> -fatty alcohol sulfate, 0.18%                   | 0                                                   |                      |
| Ex. 24 ring-opened soya polyol x 10 EO, 0.5%                              | 0                                                   |                      |

**Claims:**

1. A process for the treatment of the circulating water in painting booths, wherein the paint particles are dispersed by addition of dispersants which are selected from:

5 (a) homo- and co-polymers based on maleic acid, acrylic acid and/or methacrylic acid having molecular weights in the range between 2,500 and 500,000;

10 (b) non-ionic surfactants;

(c) anionic surfactants, no polyaspartic acid being additionally added to the circulating water in this case;

(d) inorganic or non-polymeric organic complexing agents and mixtures thereof;

15 the total concentration thereof being between 0.01 and 2.0 wt. %, based on the circulating water.

2. A process as claimed in claim 1 wherein the dispersants are non-ionic surfactants, selected from alkoxylates of fatty acids, fatty alcohols or fatty amines having 7 to 36, preferably 10 to 22, carbon atoms in the fatty alkyl group and having 5 to 100, preferably 10 to 80, alkylene oxide units.

20

3. A process as claimed in claim 1 wherein the dispersants are inorganic or non-polymeric organic complexing agents, selected from:

25 (i) organic carboxylic acids having two to ten heteroatoms, which may coordinate on metal ions, particularly from citric acid, tartaric acid, malic acid, gluconic acid, nitrilotriacetic acid, ethylenediamine tetraacetic acid, methylglycine diacetic acid;

(ii) organic phosphonic acids, particularly from 1-hydroxyethane-1,1-diphosphonic acid, aminotrimethylene phosphonic acid and phosphonobutane tricarboxylic acid;

30 (iii) oligomeric or polymeric inorganic phosphates, particularly Na triphosphate, Na pyrophosphate and Na hexametaphosphate.

4. A process as claimed in one of claims 1 to 3 wherein dispersants selected from:

5 (a) homo- and co-polymers of acrylic acid and/or methacrylic acid having molecular weights of between 2,500 and 500,000, preferably from 15,000 to 250,000;

10 (b) non-ionic surfactants;

(d) inorganic or non-polymeric organic complexing agents; are used and 0.2 to 2 wt. %, based on the circulating water, of polyaspartic acid is additionally added to the circulating water.

15 5. A process as claimed in claim 1 wherein the dispersants are anionic surfactants which are selected from soaps, alkyl sulfates, alkyl sulfonates, alkylbenzene sulfonates, alkylether sulfates each having 7 to 44, preferably 8 to 22, carbon atoms in the alkyl group, and from sulfonated maleic acid esters.

20 6. A process as claimed in one of more of claims 1 to 5 wherein the paint particles are separated from the circulating water by means of a membrane filtration.

25 7. A process for the treatment of the circulating water in painting booths, wherein the circulating water has a hardness of at least 2° German hardness and wherein the paint particles are dispersed by addition of dispersants in a concentration between 0.01 and 2.0 wt. % based on the circulating water in such a way that they exhibit an average particle size, which may be determined by means of laser diffraction, of below 20  $\mu\text{m}$ .

30 8. A process as claimed in claim 7 wherein the dispersants are selected from:

(a) homo- and co-polymers based on maleic acid, acrylic acid and/or methacrylic acid having molecular weights of between 2,500 and 500,000;

(b) non-ionic surfactants;

(d) inorganic or non-polymeric organic complexing agents and mixtures

thereof.

9. A process as claimed in claim 7 wherein the dispersants are non-ionic surfactants selected from alkoxylates of fatty acids, fatty alcohols or fatty amines having 7 to 36, preferably 10 to 22, carbon atoms in the fatty alkyl group and having 5 to 100, preferably 10 to 80, alkylene oxide units.

10. A process as claimed in claim 7 wherein the dispersants are inorganic or non-polymeric organic complexing agents selected from:

10 (i) organic carboxylic acids having two to ten heteroatoms, which may coordinate on metal ions, particularly from citric acid, tartaric acid, malic acid, gluconic acid, nitrilotriacetic acid, ethylenediamine tetraacetic acid, methylglycine diacetic acid;

15 (ii) organic phosphonic acids, particularly from 1-hydroxyethane-1,1-diphosphonic acid, aminotrimethylene phosphonic acid and phosphonobutane tricarboxylic acid;

(iii) oligomeric or polymeric inorganic phosphates, particularly Na triphosphate, Na pyrophosphate and Na hexametaphosphate.

20 11. A process as claimed in one or more of claims 7 to 10 wherein dispersants selected from:

(a) homo- and co-polymers of acrylic acid and/or methacrylic acid having molecular weights of between 2,500 and 500,000, preferably from 15,000 to 250,000;

25 (b) non-ionic surfactants;

(d) inorganic or non-polymeric organic complexing agents; are used and 0.2 to 2 wt. %, based on the circulating water, of polyaspartic acid is additionally added to the circulating water.

30 12. A process as claimed in claim 7 wherein the dispersants are selected from:

(c) anionic surfactants

and no polyaspartic acid is added to the circulating water.

13. A process as claimed in claim 12 wherein the anionic surfactants are selected from soaps, alkyl sulfates, alkyl sulfonates, alkylbenzene sulfonates, alkylether sulfates each having 7 to 44, preferably 8 to 22, carbon atoms in the alkyl group, and from sulfonated maleic acid esters.

5

14. A process as claimed in one or more of claims 7 to 13 wherein the paint particles are separated from the circulating water by means of a membrane filtration.



## DECLARATION

Page 2

I hereby claim the benefit under Title 35, United States Code §120 of any United States application(s), or §365© of any PCT international application designating the United States of America, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT international application in the manner provided by the first paragraph of Title 35, United States Code §112.1 acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations §1.56 which became available between the filing date of the prior application and the national or PCT international filing date of this application.

| U.S. Parent Application Number | PCT Parent Number | Parent Filing Date (MM/DD/YYYY) | Parent Patent Number (if applicable) |
|--------------------------------|-------------------|---------------------------------|--------------------------------------|
|                                | PCT/EP00/06291    | 7/5/2000                        |                                      |

Additional U.S. or PCT international application numbers are listed on a supplemental priority sheet attached hereto.

As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith:

|                                                                                               |                      |                  |                     |                            |
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| <input type="checkbox"/> Firm Name                                                            | <input type="text"/> | Customer Number  | or label            | <input type="text"/> 00423 |
| <input type="checkbox"/> List Attorney(s) and/or agent(s) name and registration number below: |                      |                  |                     |                            |
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I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

|                                     |                                                                               |                                                                               |  |             |                  |                     |         |
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## DECLARATION

### ADDITIONAL INVENTOR(S) Supplemental Sheet

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